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Abstract

A Reprocessing Alternative for Zircaloy Clad Natural or Slightly Enriched Uranium Oxide Fuel Elements

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The main interest for processing spent fuel elements containing natural or slightly enriched uranium is the recovery of the plutonium produced during irradiation. The recycling of depleted uranium in thermal power reactors seems at present to be of low economical value. In the reprocessing cycle it is therefore not necessary to decontaminate the uranium further than required for its storage without increasing the cost for radiation shielding.

The reprocessing method described here is intended for the treatment of uranium oxide fuel canned in zirconium alloys and irradiated to a burn-up of the order of 10,000 Mwd/t. The decanning is carried out by high temperature oxidation of the zirconium followed by a dissolution of the oxide core in nitric acid. Plutonium is separated from the uranium and fission products by an amine extraction process. The uranium is decontaminated to an adequate degree with tributyl phosphate extraction.

An economical evaluation of a plant of 1 t/day capacity is made.

1. The chemical head-end method usually proposed for Zircaloy clad fuel (the Zirflex process) produces considerable amounts of liquid intermediate radioactive wastes, which cause high storage costs. The proposed Thermox process has the advantage that only small amounts of solid waste emerges from the decanning step.

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Whole fuel element assemblies are introduced into a vertical furnace tube where the zirconium is oxidized in air and steam at 800 °C. Subsequently the uranium oxide is dissolved in dilute nitric acid. The zirconium oxide appears as solid waste in a volume one or two orders of magnitude less than the corresponding liquid Zirflex waste.

2. Tertiary amines used as solvents for extraction are characterized by a high selectivity for plutonium in its 4-valent state. This property is unchanged also after a high radiation exposure.

In this study two alternative chemical flowsheets are presented. After a primary plutonium extraction with a solvent containing 10% trilaurylamine (TLA) in tertiary butylbenzene and subsequent scrubbing, the organic plutonium stream in the first alternative passes through a silica gel column for plutonium reextraction and concentration. The plutonium eluted from this column is further purified in an anion exchange cycle.

The proposed silica gel process was tested in the laboratory with actually expected plutonium concentrations.

In the second alternative, the plutonium, after a primary extraction and scrubbing, is reextracted with acetic acid. A second plutonium extraction with 20 % TLA is followed by a direct precipitation of plutonium from the organic solution with oxalic acid.

The uranium is decontaminated in a separate tributyl phosphate cycle to a degree sufficient for subsequent handling and storing.

3. The plant layout and cost analysis are based upon the second chemical flow-sheet alternative.

Special equipment is required for the head-end process while equipment of more conventional design is assumed for the rest of the plant.

The cost estimate includes not only the main plant building but also general services and utilities required for operation of the plant.